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(54) Herbicidal Compositions

(72) Eberle, Dieter;  
Langauer, Theodor,  
Switzerland

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HERBICIDAL COMPOSITIONS

The present invention relates to flowable herbicidal compositions in the form of aqueous suspensions, which consist of at least one solid disperse phase and a continuous aqueous phase, and which contain as active ingredient at least one herbicidally active urea derivative.

The herbicidal compositions according to the invention are active-substance concentrates which are intended for the transport and storage of herbicidal preparations. These active-substance concentrates are adjusted before use, by dilution with water, to an active-substance concentration suitable for the particular purpose of application.

The method of formulating herbicidally active urea derivatives, on their own or in combination with other herbicidal active substances, for example triazine derivatives, as aqueous suspension concentrates is known. It has been possible hitherto, by the use of special anionic tenside combinations, to produce transport- and storage-stable suspension concentrates which contain a maximum of 500 g/litre of active substance. The production of stable aqueous suspensions having higher contents of active substance which, from the standpoint of economy and greater ease of application, would be desirable has so far



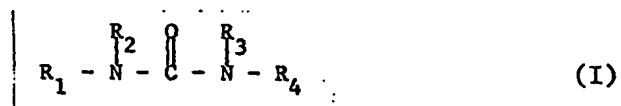
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not been possible with the tenside combinations known up to now.

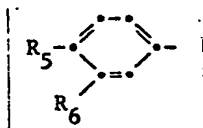
The object of the present invention therefore is to provide flowable herbicidal compositions in the form of aqueous suspensions which, even with a content of active substance of more than 500 g/litre, are stable in transport and in storage.

There is thus suggested according to the invention a flowable herbicidal suspension concentrate which contains:

35-66 % by weight of one or more urea derivatives of the formula I



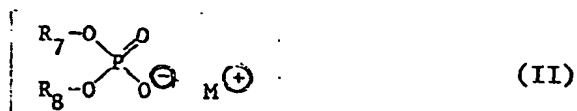
in which  $\text{R}_1$  is benzothiazol-2-yl, 5-trifluoromethyl-1,3,4-thiadiazol-2-yl or a phenyl group of the formula



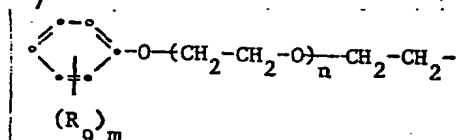
in which  $\text{R}_5$  is hydrogen, chlorine, bromine or  $\text{C}_1\text{-C}_4$ -alkyl,  $\text{R}_6$  is hydrogen, chlorine, trifluoromethyl or  $\text{C}_1\text{-C}_4$ -alkylaminocarbonyloxy, wherein however  $\text{R}_5$  and  $\text{R}_6$  cannot simultaneously be hydrogen,  $\text{R}_2$  is hydrogen or, when  $\text{R}_1$  is benzothiazol-2-yl or 5-trifluoromethyl-1,3,4-thiadiazol-2-yl,  $\text{R}_2$  is methyl;  $\text{R}_3$  is methyl, and  $\text{R}_4$  is methyl or methoxy, or, when  $\text{R}_1$  is benzothiazol-2-yl or 5-trifluoromethyl-1,3,4-thiadiazol-2-yl,  $\text{R}_4$  is hydrogen, or of a mixture of one or more urea derivatives of the formula I and a further solid herbicidal active substance difficultly soluble in water;

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1-10 % by weight of at least one mono- or di-(phenol-polyglycol ether)-phosphoric acid ester, or of a salt thereof, of the formula II

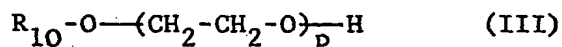


in which  $\text{R}_7$  is a radical of the formula



$\text{R}_8$  has the same meaning as  $\text{R}_7$  or is hydrogen, and  $\text{M}$  is a proton, or a sodium, potassium, diethylammonium, triethylammonium, diethanolammonium or triethanolammonium cation,  $\text{R}_9$  being hydrogen,  $\text{C}_3\text{-C}_{10}$ -alkyl or styryl,  $m$  an integer from 1-4, and  $n$  an integer from 4-40;

0.1 - 5 % by weight of at least one polyglycol ether of the formula III



wherein  $\text{R}_{10}$  is  $\text{C}_{12}\text{-C}_{18}$ -alkyl,  $\text{C}_{12}\text{-C}_{22}$ -alkanoyl,  $\text{C}_{12}\text{-C}_{22}$ -alkenoyl,  $\text{C}_{12}\text{-C}_{22}$ -alkadienonyl,  $\text{C}_{12}\text{-C}_{22}$ -alkatrienonyl, or phenyl mono- to trisubstituted by  $\text{C}_3\text{-C}_{10}$ -alkyl,  $\text{C}_5\text{-C}_8$ -cycloalkyl or styryl, and  $p$  is an integer from 1-6;

0-3 % by weight of an auxiliary dispersing agent;

0-10 % by weight of an anti-freezing agent; and

20-55 % by weight of water.

The herbicidal compositions according to the invention are suspension concentrates which contain as active

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ingredient 35-66 % by weight of one or more urea derivatives of the formula I, or of a mixture of one or more urea derivatives of the formula I with a further solid herbicide difficultly soluble in water. The suspension concentrates according to the invention preferably contain:

- 45-60 % by weight of one or more urea derivatives of the formula I, or of a mixture of one or more urea derivatives of the formula I with a further solid herbicide difficultly soluble in water;
- 2-5 % by weight of at least one mono- or di-(phenolpolyglycol ether)-phosphoric acid ester, or of a salt thereof, of the formula II;
- 0.1-2 % by weight of at least one polyglycol ether of the formula III;
- 0-3 % by weight of an auxiliary dispersing agent;
- 0-10 % by weight of an antifreezing agent; and
- 23-40 % by weight of water.

Preferred ureas of the formula I are those in which  $R_1$  is 3-chloro-4-methylphenyl, 3-trifluoromethylphenyl, 4-isopropylphenyl, 3,4-dichlorophenyl, 3-tert-butylamino-carbonyloxyphenyl, 4-bromo-3-chlorophenyl or 4-bromophenyl,  $R_2$  is hydrogen,  $R_3$  is methyl, and  $R_4$  is methyl or methoxy. Further preferred ureas of the formula I are in particular those in which  $R_1$  is benzothiazol-2-yl or 5-trifluoromethyl-1,2,4-thiadiazol-2-yl,  $R_2$  and  $R_3$  are methyl, and  $R_4$  is hydrogen. To be mentioned as especially preferred representatives of ureas of the formula I are the following:

N-(3-chloro-4-methylphenyl)-N',N'-dimethylurea (chlortoluron),  
N-(3-trifluoromethylphenyl)-N',N'-dimethylurea (fluometuron),

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N-(4-isopropylphenyl)-N',N'-dimethylurea (isoproturon),  
N-(3,4-dichlorophenyl)-N',N'-dimethylurea (diuron),  
N-(3-tert-butylaminocarbonyloxyphenyl)-N',N'-dimethylurea  
(karbutylate),  
N-(3,4-dichlorophenyl)-N'-methoxy-N'-methylurea (linuron),  
N-(4-bromo-3-chlorophenyl)-N'-methoxy-N'-methylurea  
(chlorbromuron),  
N-(4-bromophenyl)-N'-methoxy-N'-methylurea (metobromuron),  
N-(5-trifluoromethyl-1,2,4-thiadiazol-2-yl)-N,N'-dimethylurea  
(thiazafluron), and  
N-(benzothiazol-2-yl)-N,N'-dimethylurea (methabenzthiazuron).

Preferred urea derivatives of the formula I are:  
chlortoluron, metobromuron, fluometuron and isoproturon.

Further suitable herbicidal active substances which, together with one or more urea derivatives of the formula I, can form the active component of the herbicidal suspension concentrates according to the invention are essentially any solid herbicidal active substances difficultly soluble in water, so far as their combination with a urea derivative of the formula I is desirable for a particular reason, for example for broadening the range of action or to effect synergism. Such further herbicidal active substances which, together with one or more urea derivatives of the formula I, can form the active component of the suspension concentrates according to the present invention can be selected for example from the following classes of herbicidal active substances:

diphenyl ether derivatives,  
phenoxyphenoxyalkanecarboxylic acid derivatives,  
nitroanilines,  
haloacetanilides,

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O-aryloximes,  
triazines,  
benzamide derivatives,  
sulfonylureas,  
halogenated pyridyloxyalkanecarboxylic acids,  
imidazolinybenzoic acid derivatives,  
imidazolinylnicotinic acid derivatives,  
N-substituted halopyrrolidones,  
hydroxybenzonitriles and  
benzthiadiazinone derivatives.

Suitable diphenyl ether derivatives are in particular compounds of the type described in the U.S. Specification No. 3,652,645, especially 5-(2,4-dichlorophenoxy)-2-nitrobenzoic acid methyl ester (bifenox); also compounds of the type described in the German Offenlegungsschrift No. 2,831,262, particularly 2-chloro-6-nitro-3-phenoxyaniline (aclonifen); as well as compounds of the type described in the U.S. Patent Specification No. 4,322,375, especially 2-(2-chloro-4-trifluoromethylphenoxy)-5-nitrophenylphosphonic acid dimethyl ester; and compounds of the type described in the European Patent Application No. 69,055, in particular 2-[4-(3-trifluoromethylphenoxy)-phenyl]-3-ethoxycarbonyl-4-oxo-5-ethyl-5,6-dihydropyran. Suitable phenoxyphenoxy-alkanecarboxylic acid derivatives are especially compounds of the type described in the German Offenlegungsschrift No. 2,223,894, particularly  $\alpha$ -[4-(2,4-dichlorophenoxy)-phenoxy]-propionic acid methyl ester (illoxan). To be mentioned as nitroanilines are in particular compounds of the type described in the U.S. Patent Specification No. 4,199,669, preferably N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine (pendimethalin). Haloacetanilides are preferably compounds of the type described in the German Offenlegungs-



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schrift No. 2,305,495, especially N-(1-methyl-2-methoxyethyl)-N-chloroacetyl-2,6-dimethylaniline. Preferred O-aryloximes are the compounds of the type described in the U.S. Patent Specification No. 3,733,359, particularly 3,5-dibromo-4-hydroxybenzaldehyde-O-2,4-dinitrophenyloxime (bromofenoxime). Triazines which can be used are preferably compounds of the type described in the British Patent Specification No. 814,948, especially 2-ethylamino-4-tert-butylamino-6-methylthio-1,3,5-triazine (tertbutryne), 2,4-bis-isopropylamino-6-methylthio-1,3,5-triazine (prometryne) and 2,4-bis-isopropylamino-6-ethylthio-1,3,5-triazine (dipropetryn). Suitable benzamide derivatives are compounds of the type described in the European Patent Application No. 49,071, particularly N-[3-(1-ethyl-1-methylpropyl)-5-isoxazolyl]-2,6-dimethoxybenzamide (benzamizole). To be mentioned as suitable sulfonylureas are the compounds of the type described in the European Patent Application No. 44,808, especially N-[2-(2-chloroethoxy)-phenylsulfonyl]-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea, also the compounds of the type described in the U.S. Patent Specification No. 4,127,405, in particular N-(2-chlorophenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea (chlorsulfuron), and the compounds of the type described in the U.S. Patent Specification No. 4,383,113, particularly N-(2-methoxycarbonylphenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea (metsulfuron). Suitable halogenated pyridyloxyalkanecarboxylic acid derivatives are the compounds of the type described in the U.S. Patent Specification No. 4,110,104, especially 2-(4-amino-3,5-dichloro-6-fluoropyrid-2-yloxy)-acetic acid (DOWCO 433). Imidazolinybenzoic acid derivatives which are suitable are in particular the compounds of the type

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described in the U.S. Patent Specification No. 4,188,487, especially 2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-4-methylbenzoic acid methyl ester and 2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-5-methylbenzoic acid methyl ester, or a mixture of these compounds. As imidazolinylnicotinic acid derivatives, there can be used in particular the compounds described in the European Patent Application No. 41,623, especially 2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-nicotinic acid. Suitable N-substituted halopyrrolidones are the compounds described in the German Offenlegungsschrift No. 2,612,731, in particular 1-(3-trifluoromethylphenyl)-2-chloro-4-chloromethyl-2-pyrrolidone. Hydroxybenzonitriles which are suitable are preferably the compounds described in the U.S. Patent Specification No. 3,397,054, especially 3,5-dibromo-4-hydroxybenzonitrile (bromoxynil) and 3,5-diiodo-4-hydroxybenzonitrile (ioxynil). Benzthiadiazinone derivatives that can be used are preferably compounds of the type described in the German Offenlegungsschrift No. 1,542,836, particularly 3-isopropyl-(1H)-benzo-2,1,3-thiadiazin-2-one-2,2-dioxide (bentazone).

Preferred herbicidal active substances which can form, together with one or more urea derivatives of the formula I, the active component of the suspension concentrates of the invention are:

5-(2,4-dichlorophenoxy)-2-nitrobenzoic acid methyl ester,

N-(1-methyl-2-methoxyethyl)-N-chloroacetyl-2,6-dimethylaniline,

2-ethylamino-4-tert-butylamino-6-methylthio-1,3,5-triazine,

2,4-bis-isopropylamino-6-methylthio-1,3,5-triazine,

2,4-bis-isopropylamino-6-ethylthio-1,3,5-triazine,

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N-[2-(2-chloroethoxy)-phenylsulfonyl]-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea,

N-(2-chlorophenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea, and

N-(2-methoxycarbonylphenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea.

The herbicidal compositions according to the invention can contain one or more mono- or di-(phenolpolyglycol ether)-phosphoric acid esters, or salts thereof, of the formula II.

The mono- and di-(phenolpolyglycol ether)-phosphoric acid esters of the formula II are preferably used as salts, especially as triethanolammonium salts, it being possible to use the pure mono- or disalts or mixtures of mono- and disalts. As a  $C_3$ - $C_{10}$ -alkyl group, the radical  $R_9$  bound to the phenyl group of the radical  $R_7$  is in particular a straight-chain alkyl group, such as n-propyl, n-butyl, n-pentyl, n-hexyl, n-heptyl, n-octyl, n-nonyl or n-decyl; it embraces however also the branched-chain isomers of these radicals, such as tert-butyl, dimethylpentyl, dimethylhexyl, dimethyloctyl and diethylpropyl. Preferred meanings of the radical  $R_9$  are n-nonyl and styryl. The preferred meaning for m is 1-3, and the preferred meaning for n is 6-26.

Examples of substituted phenyl groups which can be present in the radical  $R_7$  are: 4-n-nonylphenyl, 2,4,6-tri-n-butylphenyl, 2,4,6-tristyrylphenyl, 2,4-di-n-nonylphenyl, 2,4,6-tri-n-pentylphenyl, 2,4-distyrylphenyl, 4-styrylphenyl, 2,3,4,6-tetrastyrylphenyl, 4-n-decylphenyl, 4-n-heptylphenyl, 4-n-pentylphenyl-2,4-di-n-hexylphenyl, 2,4-di-n-octylphenyl, 4-n-octylphenyl, 4-n-hexylphenyl, 2,4-di-n-pentylphenyl and 2,4,6-tri-n-pentylphenyl.

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Preferred phenyl groups which can be denoted by  $R_7$  or  $R_7$  and  $R_8$  are 4-n-nonylphenyl and trisubstituted phenyl groups, especially 2,4,6-tristyrylphenyl. In the mono- and di-(phenolpolyglycol ether)-phosphoric acid esters, or salts thereof, of the formula II, the meaning of n, when  $R_7$  or  $R_7$  and  $R_8$  are 4-n-nonylphenyl, is preferably 6-9, and when  $R_7$  or  $R_7$  and  $R_8$  are 2,4,6-triphenylstyryl it is preferably 16-26.

Typical representatives of mono- and di-(phenolpolyglycol ether)-phosphoric acid esters of the formula II usable according to the invention are: mono- and di-(tristyrylphenolhexadecaglycol ether)-phosphoric acid esters; mono- and di-(tristyrylphenoloctadecaglycol ether)-phosphoric acid esters, mono- and di-(tristyrylphenolnonadecaglycol ether)-phosphoric acid esters, mono- and di-(tristyrylphenoleicosaglycol ether)-phosphoric acid esters, mono- and di-(tristyrylphenoldocosaglycol ether)-phosphoric acid esters, mono- and di-(nonylphenolhexaglycol ether)-phosphoric acid esters, mono- and di-(nonylphenoloctaglycol ether)-phosphoric acid esters and mono- and di-(nonylphenolnonaglycol ether)-phosphoric acid esters. The phenolpolyglycol ether phosphoric acid esters of the formula II which are obtainable commercially are as a rule in the form of mixtures of the corresponding mono- and diesters, or as salts of mixtures of mono- and diesters. Also with regard to the substitution in the phenyl group, the products obtainable commercially are mixtures having a varying degree of substitution ( $m = 1-4$ ). Mixtures of this type can be advantageously used for producing the herbicidal compositions according to the invention.

Suitable mono- and di-(phenolpolyglycol ether)-phosphoric acid ester derivatives of the formula II which are obtainable commercially are for example:

- HOE S 3475 \* triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate, wherein the polyglycol ether moiety consists on average of 20 ethylene glycol units ( $\bar{n} = 20$ );
- Soprophor Fl \* triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate, wherein the polyglycol ether moiety contains 16-20 ethylene glycol units ( $n = 16-20$ );
- Soitem 8 FL/N\* triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate, in which the polyglycol ether moiety contains an average of 18 ethylene glycol units ( $\bar{n} = 18$ );
- Celanol PS-17\* potassium salt of a mixture of mono- and di-(nonylphenolpolyglycol ether)-ester phosphate having on average 6 ethylene glycol units ( $\bar{n} = 6$ ); and
- Celanol PS-19\* potassium salt of a mixture of mono- and di-(nonylphenolpolyglycol ether)-ester phosphate having on average 9 ethylene glycol units ( $\bar{n} = 9$ ).

The polyglycol ethers of the formula III preferably contain 1-3 glycol ether units ( $p = 1-3$ ).

As a  $C_{12}$ - $C_{18}$ -alkyl group, the radical  $R_{10}$  embraces in particular straight-chain alkyl groups, such as n-dodecyl, n-tridecyl, n-tetradecyl, n-pentadecyl, n-hexadecyl, n-heptadecyl and n-octadecyl, but also isomers thereof having branched chains, such as trimethylnonyl, tetramethylnonyl, dimethylundecyl and dipropylhexyl. As a cycloalkyl

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group having 5-8 carbon atoms,  $R_{10}$  embraces cyclopentyl, cyclohexyl, cycloheptyl and cyclooctyl.

By alkanoyl, alkenoyl, alkadienoyl and alkatrienoyl groups having 12-22 carbon atoms are meant in this case the acyl groups of the corresponding 1-, 2- or 3-fold unsaturated carboxylic acids. To be mentioned as saturated carboxylic acids from which the alkanoyl groups are derived are: lauric acid, myristic acid, palmitic acid and stearic acid. Unsaturated carboxylic acids from which the alkenyl groups denoted by  $R_{10}$  are derived are: lauroleic acid, myristoleic acid, palmitoleic acid, petroselinic acid, oleic acid, elaidic acid, vaccenic acid, ricinoleic acid and erucic acid. Examples of 2- or 3-fold unsaturated carboxylic acids from which the alkadienoyl and alkatrienoyl groups denoted by  $R_{10}$  are derived are: linoleic acid, linolenic acid, riceneic acid and  $\alpha$ -elaostearic acid.

As a mono- to trisubstituted phenyl group,  $R_{10}$  is for example 4-n-nonylphenyl, 2,4,6-tri-n-butylphenyl, 2,4,6-tristyrylphenyl, 2,4-di-n-nonylphenyl, 2,4,6-tri-n-pentylphenyl, 2,4-distyrylphenyl, 4-styrylphenyl, 4-n-decylphenyl, 4-n-heptylphenyl, 4-n-pentylphenyl, 2,4-di-n-hexylphenyl, 2,4-di-n-octylphenyl, 4-n-octylphenyl, 4-n-hexylphenyl, 2,4-di-n-pentylphenyl, 2,4,6-tri-n-pentylphenyl, 4-cyclohexylphenyl, 4-cyclopentylphenyl and 2,4-dicyclohexylphenyl.

Polyglycol ethers of the formula III obtainable commercially are for example:

Arkopal N 030\* 4-nonylphenolpolyglycol ether having on average 3 ethylene glycol units ( $\bar{p} = 3$ );

Arkopal N 040\* 4-nonylphenolpolyglycol ether having on average 4 ethylene glycol units ( $\bar{p} = 4$ );

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- Disponil NP 3<sup>\*</sup> 4-nonylphenolpolyglycol ether having on average 3 ethylene glycol units ( $\bar{p} = 3$ );
- Antarox CO 210<sup>\*</sup> 4-nonylphenolpolyglycol ether having 1-2 ethylene glycol units ( $\bar{p} = 1-2$ );
- Brij 92<sup>\*</sup> oleyl polyglycol ether having on average 2 ethylene glycol units ( $\bar{p} = 2$ ); and
- Genapol O-020<sup>\*</sup> oleyl polyglycol ether having on average 2 ethylene glycol units ( $\bar{p} = 2$ ).

Suitable auxiliary dispersing agents which can be contained in the herbicidal compositions according to the invention are both substances which favourably influence the degree of viscosity and substances which additionally stabilise the suspension. There can thus be added as auxiliary dispersing agents thickening agents soluble in water or capable of swelling in water, or synthetic or semisynthetic macromolecules. Suitable auxiliary dispersing agents which can be added to the herbicidal compositions according to the invention are: polysaccharides, especially polysaccharides of the xanthan, alginate, guar or cellulose type, or synthetic macromolecules, such as polyethylene glycols, polyvinylpyrrolidones, polyvinyl alcohols, polyacrylates, especially copolymers of acrylic acid and acrylic acid esters neutralised with triethanolamine or with alkali, or swellable, structure-forming silicates, such as pyrogenic or precipitated silicic acids, bentonites, montmorillonites, hectonites or attapulgites, or organic derivatives of aluminium silicates.

As antifreezing agents which can be added to the herbicidal compositions according to the invention to maintain flowability at low temperatures and to prevent the freezing out of water, the customary additives are suitable,

<sup>\*</sup> Trade Mark

such as ethylene glycol, propylene glycol, glycerol, di-, tri- and tetraethylene glycol and urea.

The herbicidal compositions according to the invention can contain, besides the aforementioned constituents, also customary additives, such as antifoaming agents, for example silicone oil, and preservatives, for example formaldehyde.

The herbicidal compositions according to the invention are flowable and are distinguished by good stability in transport and in storage. By virtue of the combination according to the invention of special anionic and nonionic tensides, it is possible to produce herbicidal suspension concentrates which can contain per unit volume considerably more active substance than can suspension concentrates known hitherto.

The herbicidal compositions according to the invention can be produced as follows:

The necessary amount of water is placed into a vessel with stirrer, and the tensides of the formulae II and III and optionally an auxiliary dispersing agent of the aforementioned type of synthetic polymers, an antifreezing agent and an antifoaming agents are stirred in until homogeneity of the mixture is obtained. The herbicidal active substance or the mixture of herbicidal active substances is then added; the whole is firstly homogenised for 10-30 minutes and subsequently ground fine. For fine grinding it is possible to use for example a glass-ball mill.

There can be added to the suspension concentrate thus obtained, for influencing the viscosity, a further auxiliary dispersing agent of the polysaccharide type (thickener) and if desired a preservative. For this purpose it is advantageous to stir the polysaccharide, in a separate



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vessel with stirrer, into the amount of water necessary to produce a 2.5% gel, and to then add the intended amount of preservative, for example a 37% aqueous formaldehyde solution. The mixture is stirred for 10 minutes, and afterwards allowed to swell for at least 2 hours. The resulting mixture can subsequently be added to the finely ground suspension concentrate, which has been prepared in the manner described above, in the amount required for the particular case, it being necessary here to take into account, in calculating the total amount of available water, the amount of water introduced as a result of this procedure.

Depending on the type and concentration of the active substances used, the flowable herbicidal compositions thus obtained have the following characteristics:

viscosity (20°C): 600-1200 m.Pa.s (measured in a Brookfield LV viscosimeter, measuring system III);

pH-value: 6.5 - 8.5; and

density (20°C): 1.08 - 1.3.

Some typical herbicidal compositions are given in the following Examples in order to further illustrate the present invention. These herbicidal compositions can be produced by the method described above.

Example 1: Suspension concentrate of chlortoluron

58.9 % by weight of commercial chlortoluron (99%),

3.1 % by weight of triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate having 16-20 ethylene glycol units,

0.75% by weight of nonylphenoltriglycol ether,

6.25% by weight of ethylene glycol,

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0.40 % by weight of silicone oil,  
0.12 % by weight of polysaccharide of the xantham type,  
0.40 % by weight of polyacrylic acid,  
0.12 % by weight of formaldehyde, and  
29.96 % by weight of water.

Content of active substance per litre: 700 g,  
density (20°C) : 1.19 - 1.21 g/cm<sup>3</sup>.

Example 2: Suspension concentrate of chlortoluron

66.15 % by weight of commercial chlortoluron (99%),  
3.10 % by weight of triethanolamine salt of a mixture of  
mono- and di-(2,4,6-tristyrylphenolpolyglycol  
ether)-ester phosphate having 16-20 ethylene  
glycol units,  
0.80 % by weight of nonylphenoltriglycol ether,  
6.25 % by weight of ethylene glycol,  
0.40 % by weight of silicone oil,  
0.04 % by weight of polysaccharide of the xantham type,  
0.04 % by weight of formaldehyde, and  
23.22 % by weight of water.

Content of active substance per litre: 800 g,  
density (20°C) : 1.21 - 1.23 g/cm<sup>3</sup>.

Example 3: Suspension concentrate of chlortoluron and  
benzamizole

57.7 % by weight of commercial chlortoluron (99%),  
2.4 % by weight of commercial benzamizole (91.7%),  
3.0 % by weight of triethanolamine salt of a mixture of  
mono- and di-(2,4,6-tristyrylphenolpolyglycol  
ether)-ester phosphate having 16-20 ethylene  
glycol units,  
0.8 % by weight of nonylphenoltriglycol ether,

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6.0 % by weight of ethylene glycol,  
0.5 % by weight of silicone oil,  
0.12 % by weight of polysaccharide of the xantham type,  
0.12 % by weight of formaldehyde,  
29.36 % by weight of water.

Content of active substance per litre: 700 g,  
density (20°C) : 1.18 - 1.20 g/cm<sup>3</sup>.

Example 4: Suspension concentrate of chlortoluron and  
bifenox

43.5 % by weight of commercial chlortoluron (99%),  
17.3 % by weight of commercial bifenox (99.8%),  
3.1 % by weight of triethanolamine salt of a mixture of  
mono- and di-(2,4,6-tristyrylphenolpolyglycol  
ether)-ester phosphate having 16-20 ethylene  
glycol units,  
0.8 % by weight of nonylphenoltriglycol ether,  
5.0 % by weight of ethylene glycol,  
0.5 % by weight of silicone oil,  
0.4 % by weight of polyacrylic acid,  
0.15 % by weight of polysaccharide of the xantham type,  
0.15 % by weight of formaldehyde, and  
29.1 % by weight of water.

Content of active substance per litre: 700 g,  
density (20°C) : 1.15 - 1.17 g/cm<sup>3</sup>.

Example 5: Suspension concentrate of chlortoluron and  
terbutryne

18.4 % by weight of commercial chlortoluron (99%),  
36.5 % by weight of commercial terbutryne (99.6%),  
2.5 % by weight of potassium salt of a mixture of  
mono- and di-(nonylphenolpolyglycol ether)-

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ester phosphate having 6-9 ethylene glycol units,  
 1.1 % by weight of nonylphenolmono- and -diglycol ether,  
 2.7 % by weight of ethylene glycol,  
 0.5 % by weight of silicone oil,  
 0.12 % by weight of polysaccharide of the xanthan type,  
 0.12 % by weight of formaldehyde, and  
 38.06 % by weight of water.

Content of active substance per litre: 600 g,  
 density (20°C) : 1.08 - 1.10 g/cm<sup>3</sup>.

Example 6: Suspension concentrate of chlortoluron and  
 dipropetryn

32.5 % by weight of commercial chlortoluron (99%),  
 21.8 % by weight of commercial dipropetryn (98%),  
 2.8 % of potassium salt of a mixture of mono- and  
 di-(nonylphenolpolyglycol ether)-ester phosphate  
 having 6-9 ethylene glycol units,  
 0.7 % by weight of nonylphenolmono- and -diglycol ether,  
 5.6 % by weight of ethylene glycol,  
 0.4 % by weight of silicone oil,  
 0.12 % by weight of polysaccharide of the xanthan type,  
 0.12 % by weight of formaldehyde, and  
 35.96 % by weight of water.

Content of active substance per litre: 600 g,  
 density (20°C) : 1.11 - 1.13 g/cm<sup>3</sup>.

Example 7: Suspension concentrate of chlortoluron and  
 N-[2-(2-chloroethoxy)-phenylsulfonyl]-N'-  
 (4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea

58.35 % by weight of commercial chlortoluron (99%),  
 6.1 % by weight of N-[2-(2-chloroethoxy)-phenylsulfonyl]-  
 N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea  
 (commercial) (96%),

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3.1 % by weight of triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate having 16-20 ethylene glycol units,  
0.75 % by weight of nonylphenoltriglycol ether,  
6.25 % by weight of ethylene glycol,  
0.4 % by weight of silicone oil,  
0.4 % by weight of a mixture of polyacrylic acid and polyacrylic acid ester,  
0.12 % by weight of polysaccharide of the xantham type,  
0.12 % by weight of formaldehyde, and  
24.41 % by weight of water.  
Content of active substance per litre: 700 g,  
density (20°C) : 1.19 - 1.21 g/cm<sup>3</sup>.

Example 8: Suspension concentrate of fluormeturon

59.9 % by weight of commercial fluormeturon (97.3%),  
3.1 % by weight of triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate having 16-20 ethylene glycol units,  
0.8 % by weight of nonylphenoltriglycol ether,  
5.0 % by weight of ethylene glycol,  
0.5 % by weight of silicone oil,  
0.4 % by weight of a mixture of polyacrylic acid and polyacrylic acid ester, neutralised with triethanolamine,  
0.15 % by weight of polysaccharide of the xantham type,  
0.15 % by weight of formaldehyde, and  
30.0 % by weight of water.  
Content of active substance per litre: 700 g,  
density (20°C) : 1.19 - 1.21 g/cm<sup>3</sup>.

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Example 9: Suspension concentrate of fluometuron and  
N-(1-methyl-2-methoxyethyl)-N-chloroacetyl-  
2,6-dimethylaniline

- 42.55 % by weight of commercial fluometuron (97.3%),
  - 10.55 % by weight of N-(1-methyl-2-methoxyethyl)-N-chloroacetyl-2,6-dimethylaniline (98.1%),
  - 3.1 % by weight of triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate having 16-20 ethylene glycol units,
  - 0.8 % by weight of oleylpolyglycol ether having 2-4 ethylene glycol units,
  - 5.0 % by weight of ethylene glycol,
  - 0.5 % by weight of silicone oil,
  - 0.1 % by weight of a mixture of polyacrylic acid and polyacrylic acid ester, neutralised with triethanolamine,
  - 0.1 % by weight of formaldehyde, and
  - 37.3 % by weight of water.
- Content of active substance per litre: 600 g,  
density (20°C) : 1.15 - 1.17 g/cm<sup>3</sup>.

Example 10: Suspension concentrate of metobromuron

- 53.0 % by weight of commercial metobromuron (98.65%),
- 3.5 % by weight of triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate having 16-20 ethylene glycol units,
- 0.5 % by weight of nonylphenoltriglycol ether,
- 9.0 % by weight of ethylene glycol,
- 0.3 % by weight of silicone oil,
- 0.15 % by weight of polysaccharide of the xantham type,
- 0.15 % by weight of formaldehyde, and
- 33.4 % by weight of water.

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Content of active substance per litre: 670 g,  
density (20°C) : 1.28 - 1.30 g/cm<sup>3</sup>.

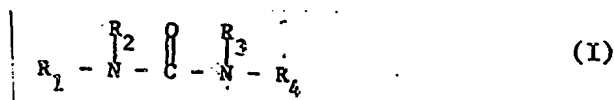
Example 11: Suspension concentrate of isoproturon

- 54.8 % by weight of commercial isoproturon (99%),
- 2.4 % by weight of triethanolamine salt of a mixture of  
mono- and di-(2,4,6-tristyrylphenolpolyglycol  
ether)-ester phosphate having 16-20 ethylene  
glycol units,
- 0.6 % by weight of oleyldiglycol ether,
- 4.0 % by weight of ethylene glycol,
- 0.5 % by weight of silicone oil,
- 0.1 % by weight of polysaccharide of the xantham type,
- 0.1 % by weight of formaldehyde,
- 37.5 % by weight of water.

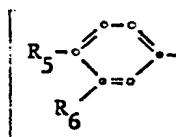
Content of active substance per litre: 600 g,  
density (20°C) : 1.08 - 1.10 g/cm<sup>3</sup>.

WHAT IS CLAIMED IS:

1. A flowable herbicidal composition in the form of an aqueous suspension which consists of at least one solid disperse phase and a continuous aqueous phase, and which contains as active component at least one herbicidally active urea derivative, which composition comprises:  
35-66 % by weight of one or more urea derivatives of the formula I



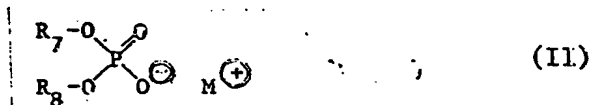
in which  $\text{R}_1$  is benzothiazol-2-yl, 5-trifluoromethyl-1,3,4-thiadiazol-2-yl or a phenyl group of the formula



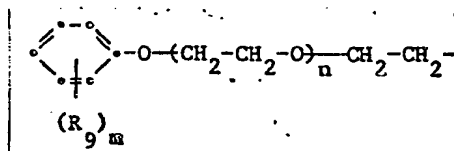
in which  $\text{R}_5$  is hydrogen, chlorine, bromine or  $\text{C}_1$ - $\text{C}_4$ -alkyl,  $\text{R}_6$  is hydrogen, chlorine, trifluoromethyl or  $\text{C}_1$ - $\text{C}_4$ -alkylaminocarbonyloxy, wherein however  $\text{R}_5$  and  $\text{R}_6$  cannot simultaneously be hydrogen,  $\text{R}_2$  is hydrogen or, when  $\text{R}_1$  is benzothiazol-2-yl or 5-trifluoromethyl-1,3,4-thiadiazol-2-yl,  $\text{R}_2$  is methyl;  $\text{R}_3$  is methyl, and  $\text{R}_4$  is methyl or methoxy, or, when  $\text{R}_1$  is benzothiazol-2-yl or 5-trifluoromethyl-1,3,4-thiadiazol-2-yl,  $\text{R}_4$  is hydrogen, or of a mixture of one or more urea derivatives of the formula I and a further solid herbicidal active substance difficultly soluble in water;



1-10 % by weight of at least one mono- or di-(phenol-polyglycol ether)-phosphoric acid ester, or of a salt thereof, of the formula II

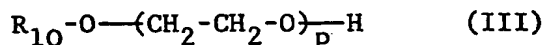


in which  $\text{R}_7$  is a radical of the formula



$\text{R}_8$  has the same meaning as  $\text{R}_7$  or is hydrogen, and  $\text{M}$  is a proton, or a sodium, potassium, diethylammonium, triethylammonium, diethanolammonium or triethanolammonium cation,  $\text{R}_9$  being hydrogen,  $\text{C}_3$ - $\text{C}_{10}$ -alkyl or styryl,  $m$  an integer from 1-4, and  $n$  an integer from 4-40;

0.1 - 5 % by weight of at least one polyglycol ether of the formula III



wherein  $\text{R}_{10}$  is  $\text{C}_{12}$ - $\text{C}_{18}$ -alkyl,  $\text{C}_{12}$ - $\text{C}_{22}$ -alkanoyl,  $\text{C}_{12}$ - $\text{C}_{22}$ -alkenoyl,  $\text{C}_{12}$ - $\text{C}_{22}$ -alkadienonyl,  $\text{C}_{12}$ - $\text{C}_{22}$ -alkatrienonyl, or phenyl mono- to trisubstituted by  $\text{C}_3$ - $\text{C}_{10}$ -alkyl,  $\text{C}_5$ - $\text{C}_8$ -cycloalkyl or styryl, and  $p$  is an integer from 1-6;

0-3 % by weight of an auxiliary dispersing agent;

0-10 % by weight of an anti-freezing agent; and

20-55 % by weight of water.

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2. A flowable herbicidal composition according to claim 1, which comprises:

45-60 % by weight of one or more urea derivatives of the formula I, or of a mixture of one or more urea derivatives of the formula I with a further solid herbicide difficultly soluble in water;

2-5 % by weight of at least one mono- or di-(phenol-polyglycol ether)-phosphoric acid ester, or of a salt thereof, of the formula II;

0.1-2 % by weight of at least one polyglycol ether of the formula III;

0-3 % by weight of an auxiliary dispersing agent;

0-10 % by weight of an anti-freezing agent; and

23-40 % by weight of water.

3. A flowable herbicidal composition according to claims 1 and 2, which contains a urea of the formula I in which  $R_1$  is 3-chloro-4-methylphenyl, 3-trifluoromethylphenyl, 4-isopropylphenyl, 3,4-dichlorophenyl, 3-tert-butylaminocarbonyloxyphenyl, 4-bromo-3-fluorophenyl or 4-bromophenyl,  $R_2$  is hydrogen,  $R_3$  is methyl, and  $R_4$  is methyl or methoxy.

4. A flowable herbicidal composition according to claims 1 and 2, which contains a urea of the formula I in which  $R_1$  is benzothiazol-2-yl or 5-trifluoromethyl-1,2,4-thiadiazol-2-yl,  $R_2$  and  $R_3$  are methyl, and  $R_4$  is hydrogen.

5. A flowable herbicidal composition according to claims 1 and 2, which contains as herbicidally active urea derivative of the formula I: chlortoluron, fluometuron,

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isoproturon, diuron, karbutylate, linuron, chlorbromuron, metobromuron, thiazafluron or methabenzthiazuron.

6. A flowable herbicidal composition according to claims 1 and 2, which contains as urea derivative of the formula I: chlortoluron, metobromuron, fluometuron or isoproturon.

7. A flowable herbicidal composition according to claim 1, which contains, besides a urea derivative of the formula I, a further solid, difficultly water-soluble, herbicidal active substance selected from the following classes of herbicidal active substances: diphenyl ether derivatives, phenoxyphenoxyalkane-carboxylic acid derivatives, nitroanilines, haloacetanilides, O-aryloximes, triazines, benzamide derivatives, sulfonylureas, halogenated pyridyl-oxyalkane-carboxylic acids, imidazolinylnicotinic acid derivatives, imidazolinylnicotinic acid derivatives, N-substituted halopyrrolidones, hydroxybenzonitriles and benzthiadiazinone derivatives.

8. A flowable herbicidal composition according to claims 1 and 7, which contains, besides a herbicidally active urea derivative of the formula I, a further solid, difficultly water-soluble, herbicidal active substance selected from the group comprising: 5-(2,4-dichlorophenoxy)-2-nitrobenzoic acid methyl ester, 2-chloro-6-nitro-3-phenoxyaniline, 2-(2-chloro-4-trifluoromethylphenoxy)-5-nitrophenylphosphonic acid dimethyl ester, 2-[4-(3-trifluoromethylphenoxy)-phenyl]-3-ethoxycarbonyl-4-oxo-5-ethyl-5,6-dihydropyran,  $\alpha$ -[4-(2,4-dichlorophenoxy)-phenoxy]-propionic acid methyl ester, N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine, N-(1-methyl-2-methoxyethyl)-N-chloroacetyl-2,6-

dimethylaniline, 3,5-dibromo-4-hydroxybenzaldehyde-0-2,4-dinitrophenyloxime, 2-ethylamino-4-tert-butylamino-6-methylthio-1,3,5-triazine, 2,4-bis-isopropylamino-6-methylthio-1,3,5-triazine, 2,4-bis-isopropylamino-6-ethylthio-1,3,5-triazine, N-[3-(1-ethyl-1-methylpropyl-5-isoxazolyl)-2,6-dimethoxybenzamide, N-[2-(2-chloroethoxy)-phenylsulfonyl]-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea, N-(2-chlorophenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea, N-(2-methoxycarbonylphenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea, 2-(4-amino-3,5-dichloro-6-fluoropyrid-2-yloxy)-acetic acid, 2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-4-methylbenzoic acid methyl ester, 2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-5-methylbenzoic acid methyl ester, 2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-nicotinic acid, 1-(3-trifluoromethylphenyl)-2-chloro-4-chloromethyl-2-pyrrolidone, 3,5-dibromo-4-hydroxybenzonitrile, 3,5-diiodo-4-hydroxybenzonitrile or 3-isopropyl-1H-benzo-2,1,3-thiadiazin-2-one-2,2-dioxide.

9. A flowable herbicidal composition according to claims 1 and 7, which contains, besides a urea derivative of the formula I, a further solid, difficultly water-soluble, herbicidal active substance selected from the group comprising: 5-(2,4-dichlorophenoxy)-2-nitrobenzoic acid methyl ester, N-(1-methyl-2-methoxyethyl)-N-chloroacetyl-2,6-dimethylaniline, 2-ethylamino-4-tert-butylamino-6-methylthio-1,3,5-triazine, 2,4-bis-isopropylamino-6-methylthio-1,3,5-triazine, 2,4-bis-isopropylamino-6-ethylthio-1,3,5-triazine, N-[2-(2-chloroethoxy)-phenylsulfonyl]-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea, N-(2-chlorophenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea and N-(2-methoxycarbonylphenylsulfonyl)-N'-(4-methoxy-6-methyl-1,3,5-triazin-2-yl)urea.

10. A flowable herbicidal composition according to claim 1, which contains, as mono- and di-(phenolpolyglycol ether)-phosphoric acid ester of the formula II, a triethanolamine salt of a mixture of a mono- and di-(phenolpolyglycol ether)-phosphoric acid ester of the formula II.
11. A flowable herbicidal composition according to claims 1 and 10, wherein mono- and di-(phenolpolyglycol ether)-phosphoric acid esters of the formula II are used, in which  $R_9$  is n-nonyl and styryl, m is 1-3, and n is 6-26.
12. A flowable herbicidal composition according to claims 1 and 2, which composition contains, as mono- and di-(phenolpolyglycol ether)-phosphoric acid esters of the formula II: mono- and di-(tristyrylphenolhexadecaglycol ether)-phosphoric acid esters, mono- and di-(tristyrylphenoloctadecaglycol ether)-phosphoric acid esters, mono- and di-(tristyrylphenolnonadecaglycol ether)-phosphoric acid esters, mono- and di-(tristyrylphenoleicosaglycol ether)-phosphoric acid esters, mono- and di-(tristyrylphenoldocosaglycol ether)-phosphoric acid esters, mono- and di-(nonylphenolhexaglycol ether)-phosphoric acid esters, mono- and di-(nonylphenoloctaglycol ether)-phosphoric acid esters and mono- and di-(nonylphenolnonaglycol ether)-phosphoric acid esters.
13. A flowable herbicidal composition according to claims 1 and 10, which composition contains, as mono- and di-(phenolpolyglycol ether)-phosphoric acid ester derivatives of the formula II, the triethanolamine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-

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ester phosphate having on average 20 ethylene glycol units in the polyglycol ether moiety, the triethanol amine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate having 16-20 ethylene glycol units in the polyglycol ether moiety, the triethanol-amine salt of a mixture of mono- and di-(2,4,6-tristyrylphenolpolyglycol ether)-ester phosphate having on average 18 ethylene glycol units in the polyglycol ether moiety, the potassium salt of a mixture of mono- and di-(nonylphenolpolyglycol ether)-ester phosphate having on average 6 ethylene glycol units in the polyglycol ether moiety, the potassium salt of a mixture of mono- and di-(nonylphenolpolyglycol ether)-ester phosphate having on average 8 ethylene glycol units in the polyglycol ether moiety or the potassium salt of a mixture of mono- and di-(nonylphenolpolyglycol ether)-ester phosphate having on average 9 ethylene glycol units in the polyglycol ether moiety.

14. A flowable herbicidal composition according to claim 1, which contains a polyglycol ether of the formula III which contains 1-3 ethylene glycol units.

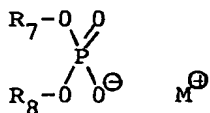
15. A flowable herbicidal composition according to claims 1 and 14, which contains, as polyglycol ether of the formula III: nonylphenolmonoglycol ether, nonylphenoldiglycol ether, nonylphenoltriglycol ether, or mixtures of such nonylphenolglycol ethers, or oleyldiglycol ether.

Fetherstonhaugh & Co.,  
Ottawa, Canada,  
Patent Agents

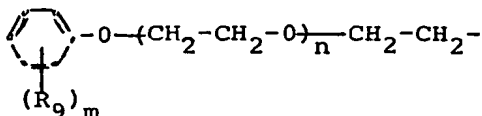


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(1)ABSTRACT

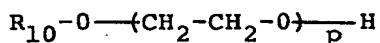
There are described flowable herbicidal compositions which contain 35-66% by weight of one or more urea derivatives, or of a mixture of one or more urea derivatives and a further solid herbicidal active substance difficultly soluble in water; 1-10% by weight of at least one mono- or di-(phenolpolyglycol ether)-phosphoric acid ester, or of a salt thereof, of the formula II



in which  $\text{R}_7$  is a radical of the formula



$\text{R}_8$  has the same meaning as  $\text{R}_7$  or is hydrogen, and  $\text{M}$  is a proton, or a sodium, potassium, diethylammonium, triethylammonium, diethanolammonium or triethanolammonium cation,  $\text{R}_9$  being hydrogen,  $\text{C}_3$ - $\text{C}_{10}$ -alkyl or styryl,  $m$  an integer from 1-4, and  $n$  an integer from 4-40; 0.1 - 5% by weight of at least one polyglycol ether of the formula III



wherein  $\text{R}_{10}$  is  $\text{C}_{12}$ - $\text{C}_{18}$ -alkyl,  $\text{C}_{12}$ - $\text{C}_{22}$ -alkanoyl,  $\text{C}_{12}$ - $\text{C}_{22}$ -alkenoyl,

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(27)

$C_{12}$ - $C_{22}$ -alkadienoyl,  $C_{12}$ - $C_{22}$ -alkatrienonyl, or phenyl mono- to trisubstituted by  $C_3$ - $C_{10}$ -alkyl,  $C_5$ - $C_8$ -cycloalkyl or styryl, and p is an integer from 1-6; 0-3% by weight of an auxiliary dispersing agent; 0-10% by weight of an anti-freezing agent; and 20-55% by weight of water.